IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
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In re the Application of: Akira YAMAGUCHI et al. Group Art Unit: 1793
Application No. 10/551,367 Examiner: Y. TAKEUCHI
Filed: November 15, 2005

For: ELECTRODE STEP DIFFERENCE ABSORBING PRINT PASTE AND METHOD OF PRODUCING ELECTRONIC DEVICE

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The Honorable Commissioner of Patents and Trademarks
United States Patent and Trademark Office
Washington, D. C. 20231

DECLARATION UNDER 37 CFR 1.132

Sir:

I, Toshio Sakurai, declare and state that:

1. In March 1994, I was graduated from Nihon University, Faculty of Humanities and Sciences and received a degree of Bachelor of Sciences from the same University. In March 1996, I was graduated from the master course of Tokyo Institute of Technology Faculty of Science, majoring in chemistry, and received a degree of Master of chemistry from the same University.

Since 1996, I have been an employee of TDK Corporation, and till the present time I have been engaged in research of mixing technique of ceramics , in research of materials of ceramics and development of the application.

2. I am familiar with the invention described in the specification of the above-identified application.

3. I carried out the following experiment. Details of my experiment are as follows.

## Experiment (comparative)

(Production of Green Sheet, Release Layer Slurry, Adhesive Layer Slurry, & Internal Electrode Paste)

The green sheet, the release layer slurry, the adhesive layer slurry and the internal electrode paste were respectively prepared according to the method disclosed in the present specification, paragraphs [0121] to [0130].

Formation of Green Sheet and Transfer of Adhesive Layer and Electrode Layer

(Production of Electrode Level Difference Absorbing Print Paste)

The same ceramic powder and subcomponent additives were prepared as with the green sheet slurry to obtain the same compounding ratio. That is, 1.48 parts by weight of $(Ba_{0.6}Ca_{0.4})SiO_3$, 1.01 parts by weight of $Y_2O_3$, 0.72 wt% of $MgCO_3$, 0.13 wt% of $Cr_2O_3$ and 0.045 wt% of $V_2O_5$ were used with respect to 100 parts by weight of the $BaTiO_3$ powder (BT-02 made by Sakai Chemical Industry Co., Ltd.).

Ceramic powder and subcomponent additives (150g) was added with a dispersant of an ester based polymer (1.5g), an imidazoline based antistatic agent (0.6g), terpineol (50g) and diotycle phthalate as a plasticizer (5g) and mixed for 4 hours. Next, the mixed solution was added with 8% lacquer (8 wt% of polyvinyl butyral and 92 wt% of terpineol with respect to the entire lacquer) of BH6 (a polyvinyl butyral resin having a polymerization degree and a butyralation degree of 69 mol% ± 3%) made by Sekisui Chemical Co., Ltd. by an amount of 120g and mixed for 16 hours. After that, 0 to 60g of terpineol was added for viscosity adjustment to produce a paste.

As shown in Table 11, electrode level difference absorbing print pastes of sample numbers 1 to 8 were produced by changing a ceramic powder content (pigment concentration /wt%) with respect to the entire paste becomes 30 to 58 wt%.

Electrode level difference absorbing print pastes of sample numbers 10 to 17, 20 to 27, 30 to 37 and 40 to 47 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 1700, 2000, 2400 and 3000 respectively as a binder resin (see Tables 12 to 15).

Electrode level difference absorbing print pastes of sample numbers 50 to 54 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 2000 as a binder resin, and making a weight ratio (resin amount) of the binder resin with respect to 100 parts by weight of the ceramic powder to 2 to 10 parts by weight (see Table 16).

Electrode level difference absorbing print pastes of sample numbers 60 to 64 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 2400 as a binder resin, and changing a butyralation degree

thereof to be in a range of 77 to 63 mol% (see Table 17).

Electrode level difference absorbing print pastes of sample numbers 70 to 74 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl acetal resin having a polymerization degree of 2400 as a binder resin, and changing an acetalization degree thereof to be in a range of 77 to 63 mol% (see Table 18).

Electrode level difference absorbing print pastes of sample numbers 80 to 86 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 2000 as a binder resin, and making dioctyl phthalate as a plasticizer contained at a ratio of 0 to 150 parts by weight with respect to 100 parts by weight of the binder resin (see Table 19).

Electrode level difference absorbing print pastes of sample numbers 90 to 94 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 2000 as a binder resin and using any one of polyethylene glycol (a hygroscopic polymer), polyalkylene glycol derivative based surfactant (amphoteric surfactant), carboxylic acid amidine salt based surfactant (amphoteric surfactant), and imidazoline based surfactant (amphoteric surfactant) as an antistatic agent, or not adding any antistatic agent (see Table 20).

(Formation of Green Sheet and Transfer of Adhesive Layer and Electrode Layer)

Further, the green sheet was formed, and the adhesive layer and the electrode layer were transferred according to the specification, paragraphs [0140] to [0145].

For all of the above samples, "hanging of paste", "stacking property (stacking precision)" and "sheet erosion" were examined as well as other properties such as viscosity of the pastes and the minimum possible printing thicknesses by the printing method (a print material thickness).

Note that the "hanging of paste" column indicates to what extent the paste hangs over the electrode from the edges. This "hanging paste" phenomenon results when a paste has a viscosity that is too low. In other words, a paste with a low viscosity is unstable, loose, and will not keep its intended figure and, for example, the paste will hang over the edges of the electrode as described in the specification, paragraph [0136]. Additionally, "stacking property (stacking precision)" and "sheet erosion" also are deteriorated when a paste has a viscosity that is too low.

From the results of the above experiment, and based on my knowledge and experience on production of electrode level difference absorbing print paste, I conclude that:

It is important to control the viscosity of the electrode level difference absorbing print paste within the claimed range.

The undersigned declares further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This 26 th day of March 2009

Respectfully submitted,

Toshio Sakurai

Toshio Sakurai

Table 11

| | Polymerization Degree | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 1 | 1450 | 6 | 30 | 69 | | 50 | Imidazolines | 4 | 0.7 | No | Good (≤ 50 μm) | Little | Easy |
| Sample 2 | 1450 | ↓ | 34 | ↓ | | ↓ | ↓ | 4 | 0.8 | ↓ | ↓ | ↓ | ↓ |
| Sample 3 | 1450 | ↓ | 38 | ↓ | | ↓ | ↓ | 5 | 1.0 | ↓ | ↓ | ↓ | ↓ |
| Sample 4 | 1450 | ↓ | 42 | ↓ | | ↓ | ↓ | 7 | 1.2 | ↓ | ↓ | ↓ | ↓ |
| Sample 5 | 1450 | ↓ | 46 | ↓ | | ↓ | ↓ | 11 | 1.6 | ↓ | ↓ | ↓ | ↓ |
| Sample 6 | 1450 | ↓ | 50 | ↓ | | ↓ | ↓ | 18 | 1.9 | ↓ | ↓ | ↓ | ↓ |
| Sample 7 | 1450 | ↓ | 54 | ↓ | | ↓ | ↓ | 30 | 2.2 | ↓ | ↓ | ↓ | ↓ |
| Sample 8 | 1450 | ↓ | 58 | ↓ | | ↓ | ↓ | 52 | | | | | |

Table 12

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 10 | 1700 | 6 | 30 | 69 | | 50 | Imidazolines | 1 | 0.7 | Yes | Bad (≥ 100 μm) | Yes | Hard |
| Sample 11 | 1700 | ↓ | 34 | ↓ | | ↓ | ↓ | 3 | 0.8 | ↓ | ↓ | ↓ | ↓ |
| Sample 12 | 1700 | ↓ | 38 | ↓ | | ↓ | ↓ | 5 | 1.0 | No | Good (≤ 50 μm) | Little | Easy |
| Sample 13 | 1700 | ↓ | 42 | ↓ | | ↓ | ↓ | 9 | 1.2 | ↓ | ↓ | ↓ | ↓ |
| Sample 14 | 1700 | ↓ | 46 | ↓ | | ↓ | ↓ | 17 | 1.6 | ↓ | ↓ | ↓ | ↓ |
| Sample 15 | 1700 | ↓ | 50 | ↓ | | ↓ | ↓ | 29 | 1.9 | ↓ | ↓ | ↓ | ↓ |
| Sample 16 | 1700 | ↓ | 54 | ↓ | | ↓ | ↓ | 45 | | | | | |
| Sample 17 | 1700 | ↓ | 58 | ↓ | | ↓ | ↓ | 70 | | | | | |

Table 13

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 20 | 2000 | 6 | 30 | 69 | | 50 | Imidazolines | 1 | 0.7 | Yes | Bad (≥ 100 μm) | Yes | Hard |
| Sample 21 | 2000 | ↓ | 34 | ↓ | | ↓ | ↓ | 4 | 0.7 | No | Good (≤ 50 μm) | Little | Easy |
| Sample 22 | 2000 | ↓ | 38 | ↓ | | ↓ | ↓ | 6 | 1.0 | No | Good (≤ 50 μm) | Little | Easy |
| Sample 23 | 2000 | ↓ | 42 | ↓ | | ↓ | ↓ | 11 | 1.2 | ↓ | ↓ | ↓ | ↓ |
| Sample 24 | 2000 | ↓ | 46 | ↓ | | ↓ | ↓ | 20 | 1.6 | ↓ | ↓ | ↓ | ↓ |
| Sample 25 | 2000 | ↓ | 50 | ↓ | | ↓ | ↓ | 37 | | | | | |
| Sample 26 | 2000 | ↓ | 54 | ↓ | | ↓ | ↓ | 64 | | | | | |
| Sample 27 | 2000 | ↓ | 58 | ↓ | | ↓ | ↓ | 90 | | | | | |

Table 14

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 30 | 2400 | 6 | 30 | 69 | | 50 | Imidazolines | 2 | 0.7 | Yes | Bad (≥ 100 μm) | Yes | Hard |
| Sample 31 | 2400 | ↓ | 34 | ↓ | | ↓ | ↓ | 5 | 0.7 | No | Good (≤ 50 μm) | Little | Easy |
| Sample 32 | 2400 | ↓ | 38 | ↓ | | ↓ | ↓ | 10 | 1.0 | No | Good (≤ 50 μm) | Little | Easy |
| Sample 33 | 2400 | ↓ | 42 | ↓ | | ↓ | ↓ | 16 | 1.2 | ↓ | ↓ | ↓ | ↓ |
| Sample 34 | 2400 | ↓ | 46 | ↓ | | ↓ | ↓ | 31 | | | | | |
| Sample 35 | 2400 | ↓ | 50 | ↓ | | ↓ | ↓ | 47 | | | | | |
| Sample 36 | 2400 | ↓ | 54 | ↓ | | ↓ | ↓ | 77 | | | | | |
| Sample 37 | 2400 | ↓ | 58 | ↓ | | ↓ | ↓ | 115 | | | | | |

Table 15

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 40 | 3000 | 6 | 30 | 69 | | 50 | Imidazolines | 4 | 0.5 | Yes | Bad (≥ 100 μm) | Yes | Hard |
| Sample 41 | 3000 | ↓ | 34 | ↓ | | ↓ | ↓ | 7 | 0.8 | No | Good (≤ 50 μm) | Little | Easy |
| Sample 42 | 3000 | ↓ | 38 | ↓ | | ↓ | ↓ | 15 | 1.1 | No | Good (≤ 50 μm) | Little | Easy |
| Sample 43 | 3000 | ↓ | 42 | ↓ | | ↓ | ↓ | 30 | | | | | |
| Sample 44 | 3000 | ↓ | 46 | ↓ | | ↓ | ↓ | 50 | | | | | |
| Sample 45 | 3000 | ↓ | 50 | ↓ | | ↓ | ↓ | 84 | | | | | |
| Sample 46 | 3000 | ↓ | 54 | ↓ | | ↓ | ↓ | 131 | | | | | |
| Sample 47 | 3000 | ↓ | 58 | ↓ | | ↓ | ↓ | 200 | | | | | |

Table 16

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet | Film Density [g/cm$^3$] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 50 | 2000 | 2 | 42 | 69 | | 50 | Imidazolines | 2 | 1.0 | Yes | Bad (≥ 100 μm) | Yes | Hard | 3.8 |
| Sample 51 | ↓ | 4 | ↓ | ↓ | | ↓ | ↓ | 4 | 1.1 | No | Good (≤ 50 μm) | Little | Easy | 3.8 |
| Sample 52 | ↓ | 6 | ↓ | ↓ | | ↓ | ↓ | 11 | 1.2 | No | Good (≤ 50 μm) | Little | Easy | 3.6 |
| Sample 53 | ↓ | 8 | ↓ | ↓ | | ↓ | ↓ | 20 | 1.3 | No | Good (≤ 50 μm) | Little | Easy | 3.4 |
| Sample 54 | ↓ | 10 | ↓ | ↓ | | ↓ | ↓ | 35 | | | | | | |

Table 17

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet | Surface Roughness Ra[μm] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 60 | 2400 | 6 | 42 | 77 | | 50 | Imidazolines | 3 | 1.2 | Yes | Bad (≥ 100 μm) | Yes | Hard | 0.55 |
| Sample 61 | ↓ | ↓ | ↓ | 74 | | ↓ | ↓ | 8 | 1.3 | No | Good (≤ 50 μm) | Little | Easy | 0.59 |
| Sample 62 | ↓ | ↓ | ↓ | 69 | | ↓ | ↓ | 16 | 1.3 | No | Good (≤ 50 μm) | Little | Easy | 0.62 |
| Sample 63 | ↓ | ↓ | ↓ | 66 | | ↓ | ↓ | 20 | 1.4 | No | Good (≤ 50 μm) | Little | Easy | 0.91 |
| Sample 64 | ↓ | ↓ | ↓ | 63 | | ↓ | ↓ | 33 | | | | | | |

Table 18

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet | Surface Roughness Ra[μm] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 70 | 2400 | 6 | 42 | | 77 | 50 | Imidazolines | 36 | | | | | | |
| Sample 71 | ↓ | ↓ | ↓ | | 74 | ↓ | ↓ | 28 | 1.4 | No | Good (≤ 50 μm) | Little | Easy | 0.59 |
| Sample 72 | ↓ | ↓ | ↓ | | 69 | ↓ | ↓ | 23 | 1.4 | No | Good (≤ 50 μm) | Little | Easy | 0.62 |
| Sample 73 | ↓ | ↓ | ↓ | | 66 | ↓ | ↓ | 14 | 1.3 | No | Good (≤ 50 μm) | Little | Easy | 0.91 |
| Sample 74 | ↓ | ↓ | ↓ | | 63 | ↓ | ↓ | 3 | 1.3 | Yes | Bad (≥ 100 μm) | Yes | Hard | 0.60 |

Table 19

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet | PET Release Force [mN/cm] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 80 | 2000 | 6 | 42 | 69 | | 0 | Imidazolines | 12 | 1.1 | No | | Little | Hard | 35 |
| Sample 81 | ↓ | ↓ | ↓ | ↓ | | 10 | ↓ | 12 | 1.1 | No | Good (≤ 50 μm) | Little | Easy | 25 |
| Sample 82 | ↓ | ↓ | ↓ | ↓ | | 30 | ↓ | 12 | 1.2 | No | Good (≤ 50 μm) | Little | Easy | 20 |
| Sample 83 | ↓ | ↓ | ↓ | ↓ | | 50 | ↓ | 12 | 1.2 | No | Good (≤ 50 μm) | Little | Easy | 16 |
| Sample 84 | ↓ | ↓ | ↓ | ↓ | | 80 | ↓ | 12 | 1.2 | No | Good (≤ 50 μm) | Little | Easy | 14.00 |
| Sample 85 | ↓ | ↓ | ↓ | ↓ | | 100 | ↓ | 11 | 1.3 | No | Good (≤ 50 μm) | Little | Easy | 10.00 |
| Sample 86 | ↓ | ↓ | ↓ | ↓ | | 150 | ↓ | 10 | 1.3 | Yes | | | | Unmeasurable |

Table 20

| | Polymerization | Resin Amt [php] | Pigment Conc. [wt%] | Butyralation Degree [mol%] | Acetalization Degree [mol%] | Plasticizer Amt [php] | Antistatic Agent Kind | Viscosity [Pa·s] (at 8[1/s]) | Print Thickness [μm] | Hanging of Paste | Stacking Property (Stacking Precision) | Sheet Erosion | Release Property of Sheet | Static Electrocity Amt [kV] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample 90 | 2000 | 6 | 42 | 69 | | 50 | *1 | 12 | 1.1 | No | Good (≤ 50 μm) | Little | Easy | 12 |
| Sample 91 | ↓ | ↓ | ↓ | ↓ | | ↓ | *2 | 12 | 1.2 | No | Good (≤ 50 μm) | Little | Easy | 11 |
| Sample 92 | ↓ | ↓ | ↓ | ↓ | | ↓ | *3 | 12 | 1.2 | No | Good (≤ 50 μm) | Little | Easy | 13 |
| Sample 93 | ↓ | ↓ | ↓ | ↓ | | ↓ | *4 | 12 | 1.2 | No | Good (≤ 30 μm) | Little | Easiest | 4 |
| Sample 94 | ↓ | ↓ | ↓ | ↓ | | ↓ | None | 12 | 1.2 | No | Bad (≥ 100 μm) | Little | Hard | 36.00 |

*1 polyethylene glycol
*2 polyalkylene glycol derivative based surfactant
*3 carboxylic acid amidine salt based surfactant
*4 imidazoline based surfactant